Thermal Conductivity and Thermal Diffusivity of Solid UO_2

Preliminary Recommendation

The recommended equation for the thermal conductivity of 95% dense solid UO_2 consists of a lattice term and a term suggested by Ronchi et al. [1] to represent the small-polaron ambipolar contribution to the thermal conductivity. The lattice term was determined by a least squares fit to the lattice contributions to the thermal conductivity obtained from thermal diffusivity measurements by Ronchi et al. [1], Hobson et al.[2], Bates [3], the Battelle Memorial Institute [4], and Los Alamos Scientific Laboratory [4], and from thermal conductivity measurements by Godfrey et al. [5] and the GE-Nuclear Systems Programs [4]. The recommended equation is

$$\lambda = \frac{100}{7.5408 + 17.692t + 3.6142t^2} + \frac{6400}{t^{5/2}} \exp\left(\frac{-16.35}{t}\right) \qquad \textbf{(1)}$$

where t is T/1000, T is in K, and λ is the thermal conductivity for 95% dense UO_2 in $W \cdot m^{-1} \cdot K^{-1}$. Figure 1 compares the recommended values for the thermal conductivity for 95% dense UO_2 with the thermal conductivity data [1-5] used in the determination of the lattice term.

Thermal conductivity values for theoretically dense UO_2 or for a different density may be calculated using the porosity relation derived by Brandt and Neurer [6], which is

$$\lambda_0 = \frac{\lambda_p}{[1 - \alpha p]}, \qquad \textbf{(2)}$$

$$\alpha = 2.6 - 0.5t$$

where t is T/1000, T is in K, p is the porosity fraction, λ_p is the thermal conductivity of UO_2 with porosity p, λ_0 is the thermal conductivity of fully dense UO_2 (i.e. porosity = 0). Values for the thermal conductivity for 95% dense UO_2 calculated from Eq.(1) and for theoretically dense UO_2 determined from Eqs.(1-2) are given in Table 1.

Uncertainty

Uncertainties have been determined from the scatter in the available data and the deviations of the data from the recommended equation. From 298 to 2000 K, the uncertainty is 10%. From 2000 to 3120 K, the uncertainty has been increased to 20% because of the large discrepancies between measurements by different investigators. Uncertainties have been included in Figure 1. Most of the data included in Figure 1 fall within these uncertainty limits. However, some of the low-temperature LASL data, which are significantly lower than other data, are below the lower 10% uncertainty. In addition, some of the data of Bates, which show considerable scatter, are outside the 10% uncertainty.

Discussion

Data for the thermal diffusivity [1-4, 7-8] and thermal conductivity [4, 5, 9] of solid UO_2 have been reassessed for the following reasons. (1) The 2000-2900 K thermal diffusivity data of Ronchi et al.[1] indicate that the high-temperature thermal diffusivity values reported by Weilbacher [7,8], which were the main high-temperature data available prior to 1999, are high. (2) Advances in understanding the heat transport mechanisms in UO_2 have led to improvements in physically-based thermal conductivity equations [1, 10-11] in which only the coefficients of the lattice contribution are determined from the thermal conductivity data. (3) Above 2670 K, heat capacity values [12 -14] previously used for conversion of thermal diffusivty data to thermal conductivity are inconsistent with recent high-temperature heat capacity measurements of Ronchi et al. [1]. High-temperature thermal conductivities calculated from thermal diffusivity data using the new heat capacity data have a different temperature dependence than values used in older assessments.

Data included in this reassessment are listed in Table 2, which also gives the percent of theoretical density of the samples, the temperature range of the measurements, and the number of data obtained for each set of measurements. Although Conway and Feith [4] report results of the General Electric (GE) Nuclear Systems Programs (NSP) thermal diffusivity measurements from 600 to 1700 K as well as data from the GE-NSP "round robin" thermal conductivity measurements, only the thermal conductivity data have been included in this assessment because comparison of the thermal diffusivity data with other data show large disagreement above 1200 K. Temperatures for the data of Stora et al. [9] and the data of Godfrey et al. [5] have been converted from the 1948 International Practical Temperature Scale (IPTS) to the 1968 IPTS.

The differences between the thermal diffusivity values of Weilbacher [7] and of Ronchi et al. [1] are clearly shown in Figure 2, which plots the inverse of measured thermal diffusivities [1-4,7] as a function of temperature. The percent of theoretical density of the samples for each set of measurements has been included in the figure legend. From 300 to 2000 K, all the inverse thermal diffusivity data show a linear dependence on temperature. Although the data of Hobson et al. [2] and that of Ronchi et al. [1] continue to increase linearly with temperature to 2400 K, values from the measurements of Weilbacher deviate from the linear dependence above 2000 K. Ronchi et al.[1] attribute the high diffusivity values obtained by Weilbacher to incorrect determinations of the temperature rise of the front of the sample and to errors in the Cowan correction during data reduction. Measurements of Bates [3] on three different samples span almost the entire temperature range but show considerable scatter. At low temperatures, values of the inverse thermal diffusivity from Bates' measurements are below the values of Ronchi et al. Between 2000 and 2400 K, Bates' values fall between Weilbacher's values and those of Ronchi et al. However, the highest temperature datum of Bates is consistent with the data of Ronchi et al.

Since 1981, theoretical research and new measurements have led to improvements in equations for the thermal conductivity of UO_2. The physically-based equation of Hyland [10]

included lattice, radiation, and ambipolar contributions. The equation of Hardin and Martin [11], which was recommended in the previous INSC assessment [15] consisted of a lattice term and a small-polaron ambipolar contribution. Since the publication of these equations, Casado, Harding, and Hyland [16] have shown that the temperature dependence used by Killeen [17] in analysis of his electrical conductivity data is incorrect. This temperature dependence had been incorporated in the small-polaron ambipolar contribution in the thermal conductivity equations of Hyland [10] and of Harding and Martin [11]. Casado et al. [16] report that the correct temperature dependence for the small polaron contribution to the direct current electrical conductivity, σ (T), is

$$\sigma(T) = \frac{\sigma_1}{T^{3/2}} e^{-\varepsilon/kT} \quad \textbf{(3)}$$

where ε is the activation energy in eV of the direct current electrical conductivity, σ_1 , k is the Boltzmann constant, and T is the temperature. Ronchi et al. [1] have used this temperature dependence to refit the electrical conductivity data of Killeen [17] and determine a new term for the small-polaron ambipolar contribution to the thermal conductivity of UO_2. They determined the lattice contribution by fitting the thermal resistivities obtained from their thermal diffusivity measurements from 550 to 1100 K. They concluded that any radiative contribution to the thermal conductivity of solid UO_2 would be insignificant compared to the lattice and small-polaron ambipolar contributions. The equation given by Ronchi et al. for the thermal conductivity of 95% dense UO_2 is:

$$\lambda = \frac{100}{6.548 + 23.533t} + \frac{6400}{t^{5/2}} \exp\left(\frac{-16.35}{t}\right) \quad \textbf{(4)}$$

where t is T/1000, T is in K, and λ is the thermal conductivity for 95% dense UO_2 in $W \cdot m^{-1} \cdot K^{-1}$. The first term of this equation is the lattice contribution; the second term is the small-polaron ambipolar contribution.

Ronchi et al.[1] also fit their data to the polynomial

$$\lambda = +12.57829 - 2.31100\text{x}10^{-2}T + 2.36675\text{x}10^{-5}T^2 - 1.30812\text{x}10^{-8}T^3 + 3.63730\text{x}10^{-12}T^4 - 3.90508\text{x}10^{-16}T^5 \quad \textbf{(5)}$$

where λ is the thermal conductivity for 95% dense UO_2 in $W \cdot m^{-1} \cdot K^{-1}$ and T is the temperature in K.

In Figure 3, these two equations of Ronchi et al. are compared with the data listed in Table 2 converted to thermal conductivity for 95% dense UO_2. Thermal conductivities have been calculated from thermal diffusivity measurements [2-4,7-8] using the relationship

$$\lambda = D\ \rho\ C_P \quad \textbf{(6)}$$

where λ is the thermal conductivity, D is the measured thermal diffusivity, ρ is the sample density and C_p is the heat capacity. The sample density at temperature T, was calculated using

$$\rho(T) = F\ \rho(273) \left(\frac{L_{273}}{L_T} \right)^3 \quad \textbf{(7)}$$

where F is the fraction of theoretical density, ρ (273) is the theroretical density at 273 K = 10.963 $Mg \cdot m^{-3}$, and the ratio of L_{273}/L_T as a function of temperature is given by the equations of Martin [18], For 273 K $\leq$ T $\leq$ 923 K,

$$L_T = L_{273}\ (9.9734\text{x}10^{-1} + 9.802\text{x}10^{-6}T - 2.705\text{x}10^{-10}T^2 + 4.391\text{x}10^{-13}T^3); \quad \textbf{(8)}$$

for 923 K $\leq$ T $\leq$ 3120 K,

$$L_T = L_{273}\,(9.9672\text{x}10^{-1} + 1.179\text{x}10^{-5}T - 2.429\text{x}10^{-9}T^2 + 1.219\text{x}10^{-12}T^3) \quad \textbf{(9)}$$

where L_T and L_{273} are the lengths at temperatures T and 273 K, respectively. The heat capacity, C_p, was calculated from an equation developed by Fink [19] based on a combined analysis of enthalpy and heat capacity data that included the new heat capacity data of Ronchi et al. [1].

For 298.15 K $\leq$ T $\leq$ 3120 K

$$C_P = \frac{C_1\theta^2 e^{\theta/T}}{T^2(e^{\theta/T}-1)^2} + 2C_2T + \frac{C_3E_a e^{-E_a/T}}{T^2} \quad \textbf{(10)}$$

where C_1 = 81.613,

θ = 548.68,

$C_2 = 2.285 \times 10^{-3}$,

$C_3 = 2.360 \times 10^{7}$,

E_a = 18531.7,

T is the temperature in K, and the heat capacity, C_p, is in $J \cdot mol^{-1} \cdot K^{-1}$. All thermal conductivities were converted to 95% theoretically dense UO_2 using Eq.(2), the equation recommended by Brandt and Neuer [6].

For the thermal diffusivity measurements of Ronchi et al.[1], the values of the thermal conductivities tabulated in their paper have been used in this evaluation because these values obtained from the simultaneous measurements of thermal diffusivity and heat capacity have a higher degree of confidence than values obtained using an equation that fits the heat capacity data but does

not exactly reproduce experimental values at any given temperature.

Figure 3 shows that the high-temperature thermal conductivities of Stora and the "round robin" Grenoble data are high compared to the equation suggested by Ronchi et al. These thermal conductivity data were obtained by the radial heat flow method. Ronchi et al. question the reliablity of the high-temperature data of Stora because of vaporization of the sample and mechanical deformations above 2500 K. From 2625 to 2657 K, the GE-NSP data show significant scatter. Conway and Feith [4] state that these data should be treated with caution because examination of the GE-NSP samples following high-temperature radial heat flow measurements showed evaporation from the center of the disc and deposition of condensed material along the cooler edges. These questionable data are of the same magnitude or higher than the thermal conductivities obtained from the thermal diffusivity measurements of Weilbacher, which were questioned by Ronchi et al.[1].

Comparison of Eq.(4) with the data, shows that although it appears low relative to the lowest temperature data, it is high relative to the minimum near 2000 K. This might be attributed to the linear temperature dependence of the lattice term, which includes only constant volume three-phonon scattering processes. In their determination of the lattice term from their low temperature data (560-1100 K), Ronchi et al. considered including a quadratic temperature term to account for constant pressure thermal expansion contributions but the additional term was not statistically justified. In an attempt to improve agreement at low temperatures (below 550 K) and in the region of the thermal conductivity minimum, the lattice contribution has been reexamined.

The lattice term has traditionally been determined by fitting the low-temperature thermal conductivity data because the lattice contribution dominates the thermal conductivity at low temperatures. Figure 4 shows the total thermal conductivity, the lattice contribution, and the ambipolar contribution as a function of temperature that have been calculated from the equation of Ronchi et al., eq.(4). Below 1300 K, the ambipolar term is insignificant and the total thermal

conductivity equals the lattice contribution. Although the ambipolar term begins to have a significant contribution to the total thermal conductivity above 1300 K, it is not larger than the lattice contribution determined by Ronchi et al. until 2800 K. Even at 3120 K, the lattice contribution is still significant. Because Ronchi et al. have developed a theoretically-based term for the ambipolar contribution, which is independent of the thermal conductivity data, it is now possible to determine the lattice contribution for the entire temperature range by subtracting the ambipolar contribution given by Ronchi et al. from the experimentally determined total thermal conductivities. Because the data of Weilbacher, the data of Stora, the Grenoble data and the GE-NSP data above 2600 K are questionable, they have not been included in this analysis.

The lattice contribution to the thermal conductivity was determined by subtracting the ambipolar contribution calculated from the equation of Ronchi et al., Eq.(4), from the thermal conductivities from the measurements of Ronchi et al. [1], Hobson et al.[2], Bates [3], the Battelle Memorial Institute [4], the Los Alamos Scientific Laboratory [4], Godfrey et al. [5] and the GE-NSP. Only the GE-NSP data below 2600 K have been used. The inverse of the lattice contributions were fit to equations with both linear and quadratic temperature dependence. Figure 5 shows a comparison of the linear and quadratic fits to the inverse of the lattice contribution to the thermal conductivity. Goodness of fit tests indicated that a quadratic term is justified for this larger set of data, which spans the entire temperature region. In Figure 6, the data for the total thermal conductivity considered in this analysis are compared with (1) an equation consisting of a lattice term that is linear in temperature and the small-polaron ambipolar term given by Ronchi et al. and with (2) an equation consisting of a lattice term with quadratic temperature dependence and the small-polaron ambipolar term given by Ronchi et al. Figures 7 and 8 show the percent deviations of these equations from the data defined as:

$$\%\ Deviation = \frac{(Data - Equation)}{Data} \cdot 100\% \qquad \textbf{(11)}$$

The percent deviations from the equation with the new linear lattice term are skewed with respect to temperature and the deviations are greater than the deviations for the equation with the quadratic lattice term. These deviation plots confirm the statistical analysis that indicates that the quadratic temperature term is justified.

The recommended equation for the thermal conductivity of 95% dense UO_2, Eq.(1), includes the quadratic lattice term and the small-polaron ambipolar contribution determined by Ronchi et al.[1]. Figure 9 shows the data fit, the recommended equation, Eq.(1), the equation of Ronchi et al., Eq.(4), and the polynomial fit by Ronchi et al to their data, Eq.(5). The recommended equation fits the data near the thermal conductivity minimum and the low-temperature data of Bates better than Eq.(4). At intermediate and high temperatures, the recommended equation is very close to the polynomial fit to the data of Ronchi et al. Figure 10 shows the percent deviations of the equation of Ronchi et al., Eq.(4), from the data included in the above analysis. The percent deviations are skewed similar to those in Figure 7 for the other equation with a linear lattice term. Comparison of percent deviations in Figure 10 and Figure 8 indicates that, in general, the deviations from Eq.(4) are larger than the deviations from the recommended equation. The recommended equation fits the data of Ronchi et al., Bates, Hobson et al., Godfrey et al., and the 'round robin' data from BMI, LASL, and GE-NSP below 2600 K with a percent standard deviation of 6.2%. The standard deviation of these data from the equation given by Ronchi et al., Eq.(4), is 7.9%. Table 2 lists the percent standard deviations from Eq. (1) and from Eq.(4), the equation of Ronchi et al., for each set of data.

From their research, Ronchi et al. concluded that the solid thermal conductivity of 95% dense UO_2 at the melting point, T_m, should be in the range $2.4 \leq \lambda(T_m) \leq 3.1$ $W \cdot m^{-1} \cdot K^{-1}$. The thermal conductivity for 95% dense UO_2 at 3120 K calculated with the recommended equation, Eq.(1), is 3.0 $W \cdot m^{-1} \cdot K^{-1}$, which is consistent with the conclusion of Ronchi et al. For theoretically dense UO_2, the thermal conductivity at 3120 K calculated from Eq.(1) is 3.2 $W \cdot m^{-1} \cdot K^{-1}$. The equation given by Harding and Martin [11], which was previously recommended, in ANL/RE-97/2 [15], gave

a melting point thermal conductivity of theoretically dense solid UO_2 of 4.0 $W \cdot m^{-1} \cdot K^{-1}$, which is 3.8 $W \cdot m^{-1} \cdot K^{-1}$ for 95% dense UO_2. In Figure 11, the recommended values for the thermal conductivity of 95% dense UO_2 are compared with values from the equation of Harding and Martin. The data used in this analysis as well as the excluded data of Stora, Weilbacher, Grenoble, and GE-NSP that had been included in past assessments are shown in Figure 11. The higher melting point thermal conductivity given by the equation of Harding and Martin is consistent with the data of Weilbacher but not with the new measurements of Ronchi et al. Percent deviations of the recommended equation from this larger set of data are shown in Figure 12. Figure 12 shows that most of the percent deviations for this larger set of data fall within the recommended uncertainties (10% below 2000 K; 20% above 2000 K).

In Figure 13, the recommended values for the thermal conductivity of fully dense UO_2 are compared with other equations for the thermal conductivity of theoretically dense UO_2. Equations included in this comparison are the equation by Ronchi et al. [1], the equation given in the MATPRO database for SCADAP/RELAP5 [14], and the equation of Harding and Martin [15], which had been previously recommended in ANL/RE-97/2. Percent deviations of values calculated with these equations from the recommended values defined as

$$\%\ Deviation = \frac{(Other - Recommended)}{Recommended} \cdot 100\% \qquad \textbf{(12)}$$

are shown in Figure 14. Most of the deviations are within the uncertainties. However the percent deviations for the equation of Harding and Martin are greater than 20% above 2800 K.

Historically, the paucity of high-temperature thermal conductivity data prompted the practice of comparing thermal conductivity equations to the in-reactor conductivity integral to melt (CIM) defined as:

$$CIM = \int_{773K}^{T_m} \lambda(T)\, dT \qquad \textbf{(13)}$$

where $\lambda(T)$ is the thermal conductivity at temperature T and T_m is the melting point. This integral represents the reactor linear power at which melting begins on the centerline of a fuel pellet whose outer surface is assumed to be at 773 K. The CIM obtained from the recommended equation, Eq.(1), is 6.09 kW · m^{-1}. The polynomial used by Ronchi et al. to fit their data gives a CIM of 6.08 kW · m^{-1}. Experimental values for CIM range from 5.5 to 7.5 kW · m^{-1}. Because in-reactor CIM measurements are subject to systematic errors such as determination of the pellet surface temperature from the cladding temperature and the fuel-cladding gap conductance, and considerable controversy exists in interpretation of the melt boundary from the post-test metallurgical examinations, the CIM value is still uncertain. However, CIM values near 6.8 kW · m^{-1} have been recommended for 95% dense fuel [20]. These values were consistent with equations [10,11] that gave good agreement with the high-temperature thermal conductivity of Weilbacher. Ronchi et al.[1] state that although the most complete set of measurements at GE-San Jose' gave 6.3 $\pm$ 0.3 kW · m^{-1} for CIM [21], these results were not accepted because they were below values based on laboratory thermal conductivity and thermal diffusivity measurements. The GE values and the previous recommendations should be reconsidered now that more reliable laboratory data are available at high temperatures.

References

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Table 1. Thermal Conductivity of UO_2 with 95% and 100% Density

Temperature K	Thermal Conductivity W m^{-1} K^{-1}	
	95% Dense	100% Dense
298.15	7.61	8.68
300	7.59	8.65
400	6.58	7.48
500	5.78	6.55
600	5.14	5.81
700	4.61	5.19
800	4.17	4.68
900	3.79	4.25
1000	3.47	3.87
1100	3.19	3.55
1200	2.95	3.28
1300	2.74	3.04
1400	2.56	2.83
1500	2.41	2.66
1600	2.29	2.52
1700	2.19	2.40
1800	2.12	2.32
1900	2.08	2.27
2000	2.06	2.24
2100	2.07	2.24
2200	2.09	2.26
2300	2.14	2.30
2400	2.20	2.37
2500	2.28	2.45
2600	2.37	2.54
2700	2.48	2.64
2800	2.59	2.76
2900	2.71	2.88
3000	2.84	3.00
3100	2.97	3.13
3120	2.99	3.16

Table 2 Standard Deviations of Data from Thermal Conductivity Equations

Data Reference	Percent of Theoretical Density, %	Temperature Range, K	# of Data	Standard Deviation, %	
				Ronchi Eq.(4)	Recommended Eq.(1)
Thermal Diffusivity Measurements					
Ronchi et al. 1999 [1]	95	550 - 1100 2000-2900	125	8.6	7.2
Hobson et al. 1974 [2]	95	537 - 2488	34	8.0	3.6
Bates (3 samples) 1970 [3]	98.4	289 - 2777	188	8.7	6.0
Battelle Memorial Institute, 1969 [4]	97.4	457 - 2271	27	6.8	4.2
Los Alamos Scientific Lab. 1969 [4]	98	299 - 2083	35	9.7	12.4
Weilbacher (2 runs) 1972 [7,8]	98	773 - 3023	32	7.1	11.4
Thermal Conductivity Measurements					
GE Nuclear Systems Programs, 1969 [4]	98	1229 - 2661	70	5.9	8.0
Godfrey 1964 [5]	93.4	323 - 1573	105	7.5	3.7
Centre d'Etudes Nucleaires (CEN) Grenoble, 1969 [4]	97	373 - 2577	14	8.0	10.6
Stora 1964 [9]	95	473 - 2777	19	8.4	10.9

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Figure 1 Thermal Conductivity of 95% Dense UO_2

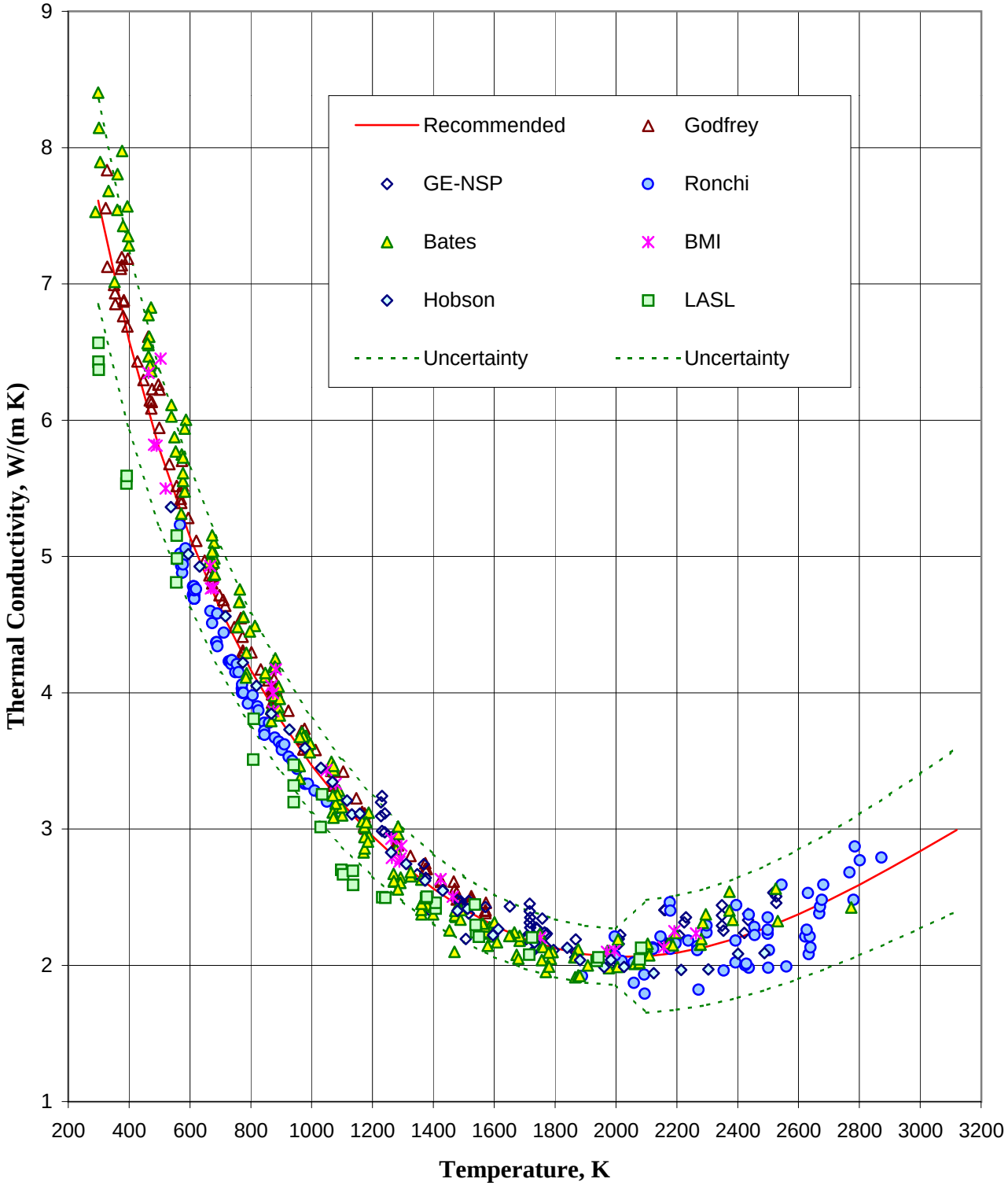

Figure 2 Inverse Thermal Diffusivity of UO_2

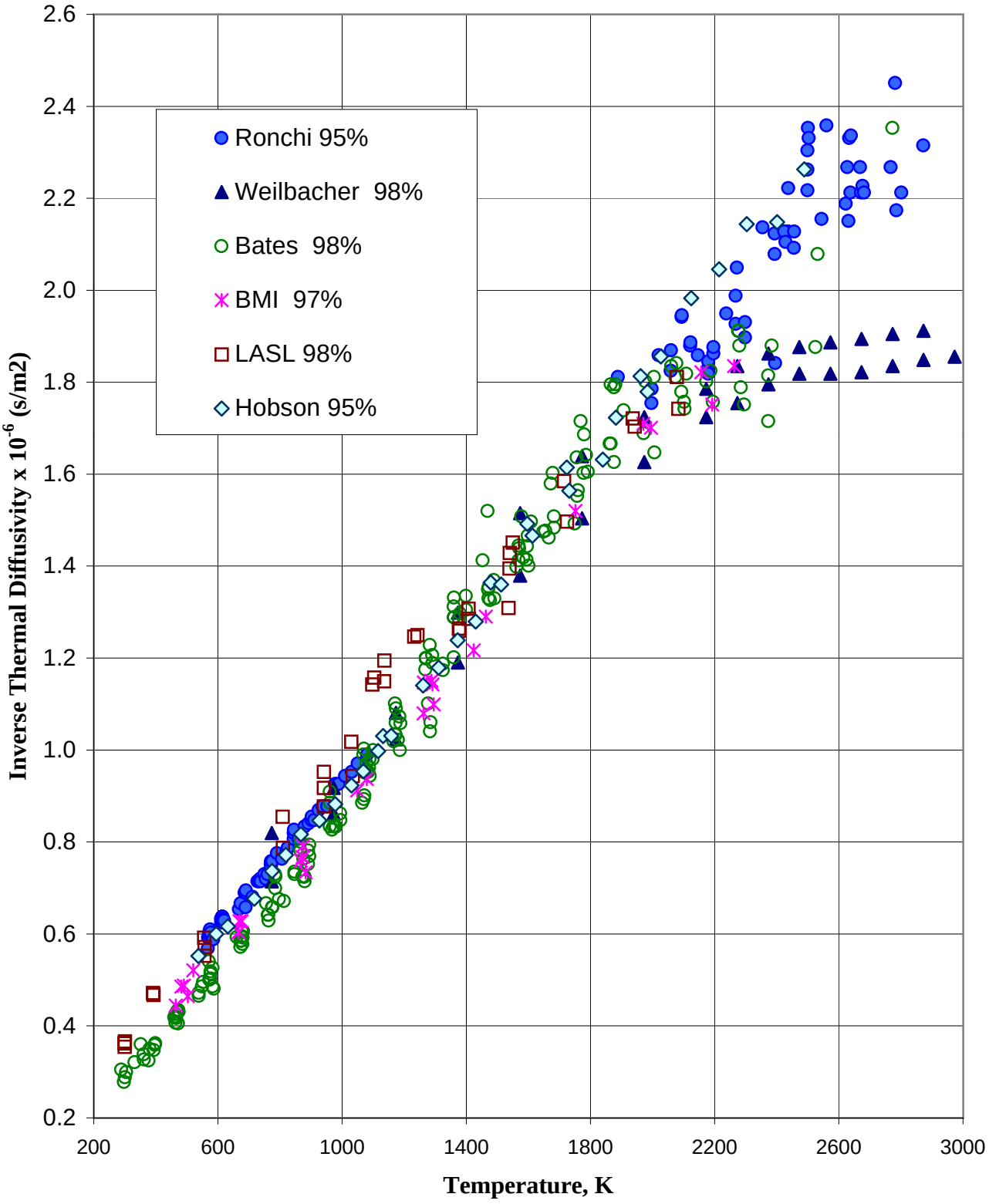

Figure 3 Equations of Ronchi et al. for the Thermal Conductivity of 95% Dense UO_2

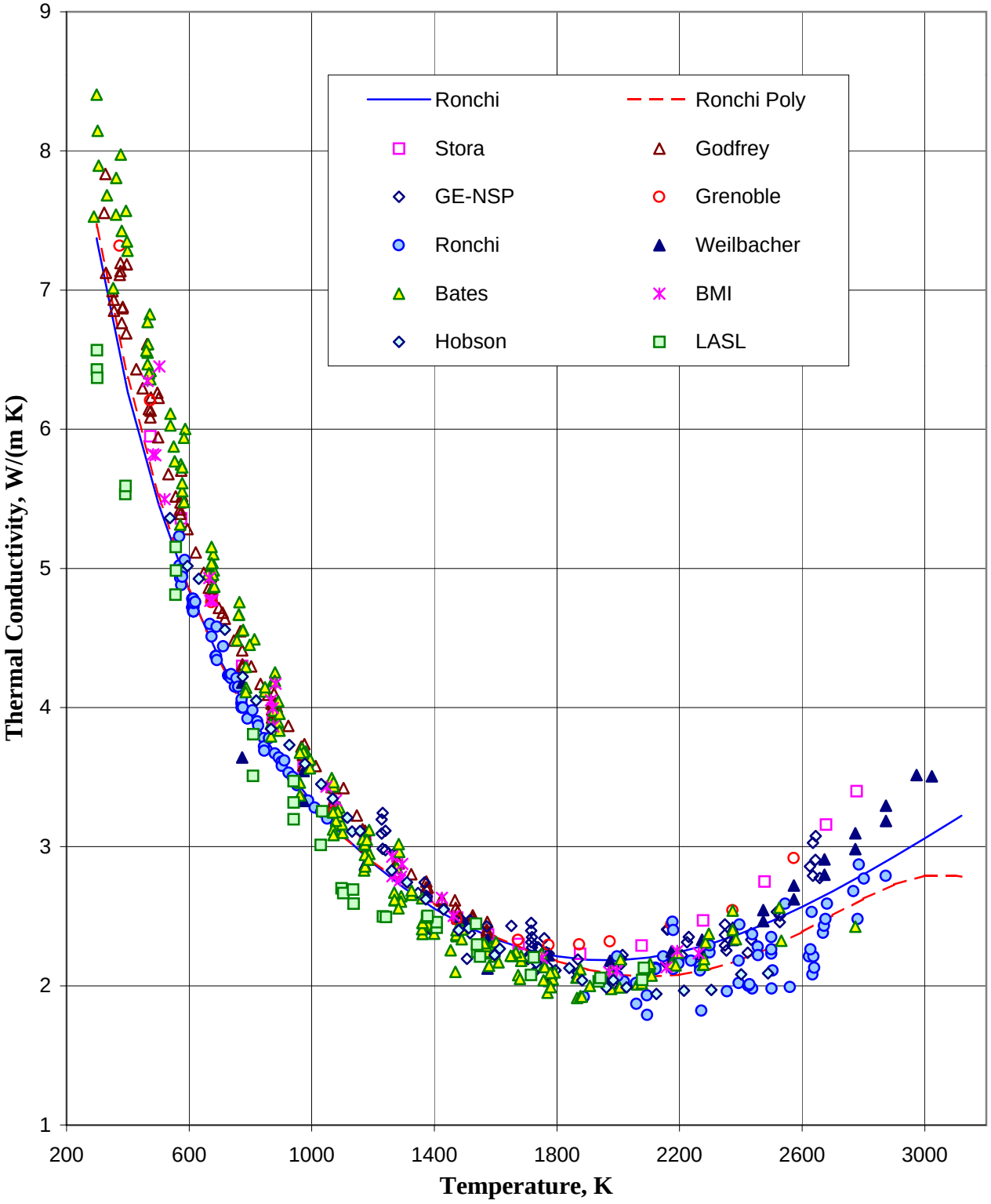

Figure 4 Thermal Conductivity Contributions from the Equation of Ronchi et al.

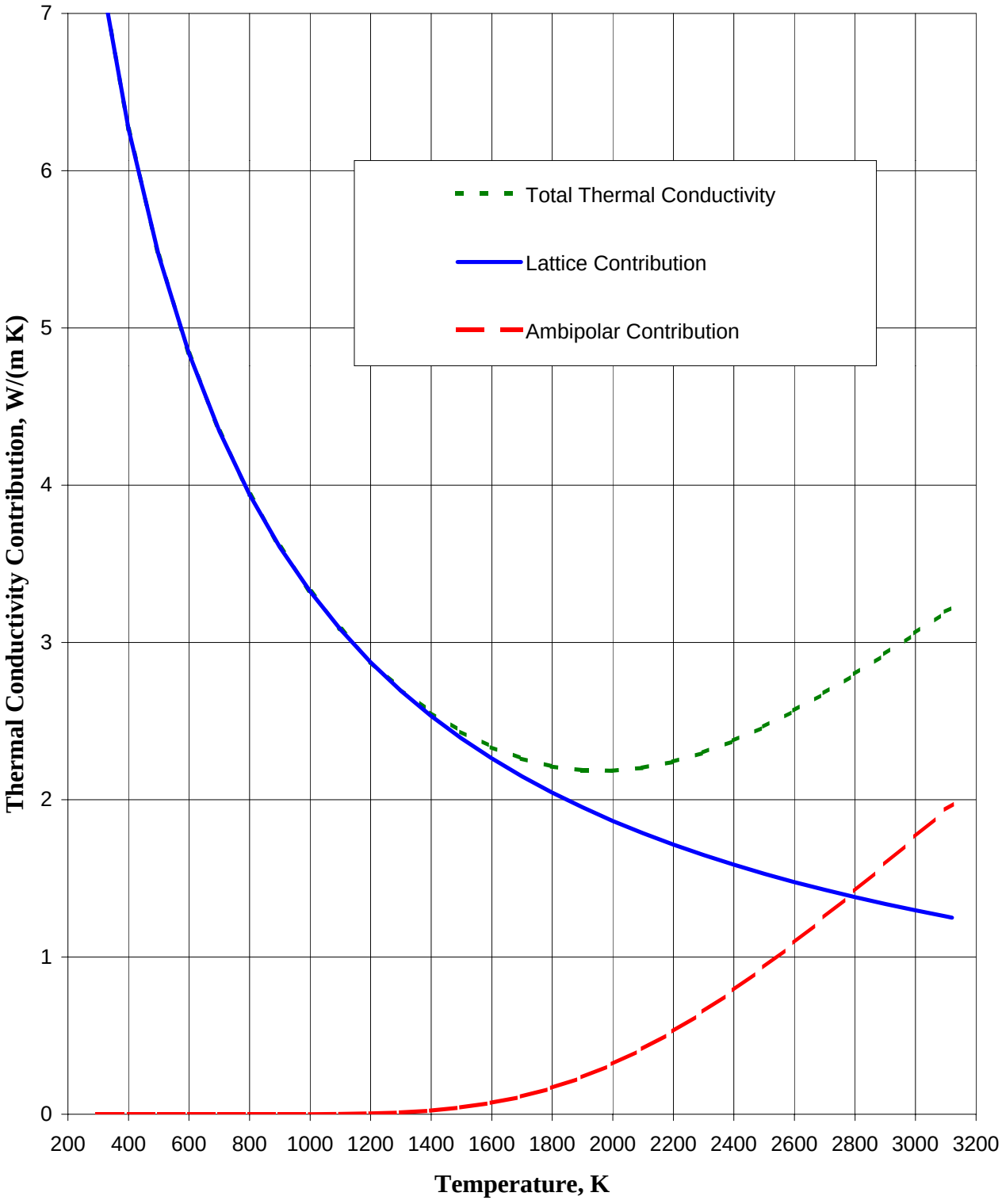

Figure 5 Linear and Quadratic Fits to Inverse Lattice Contributions

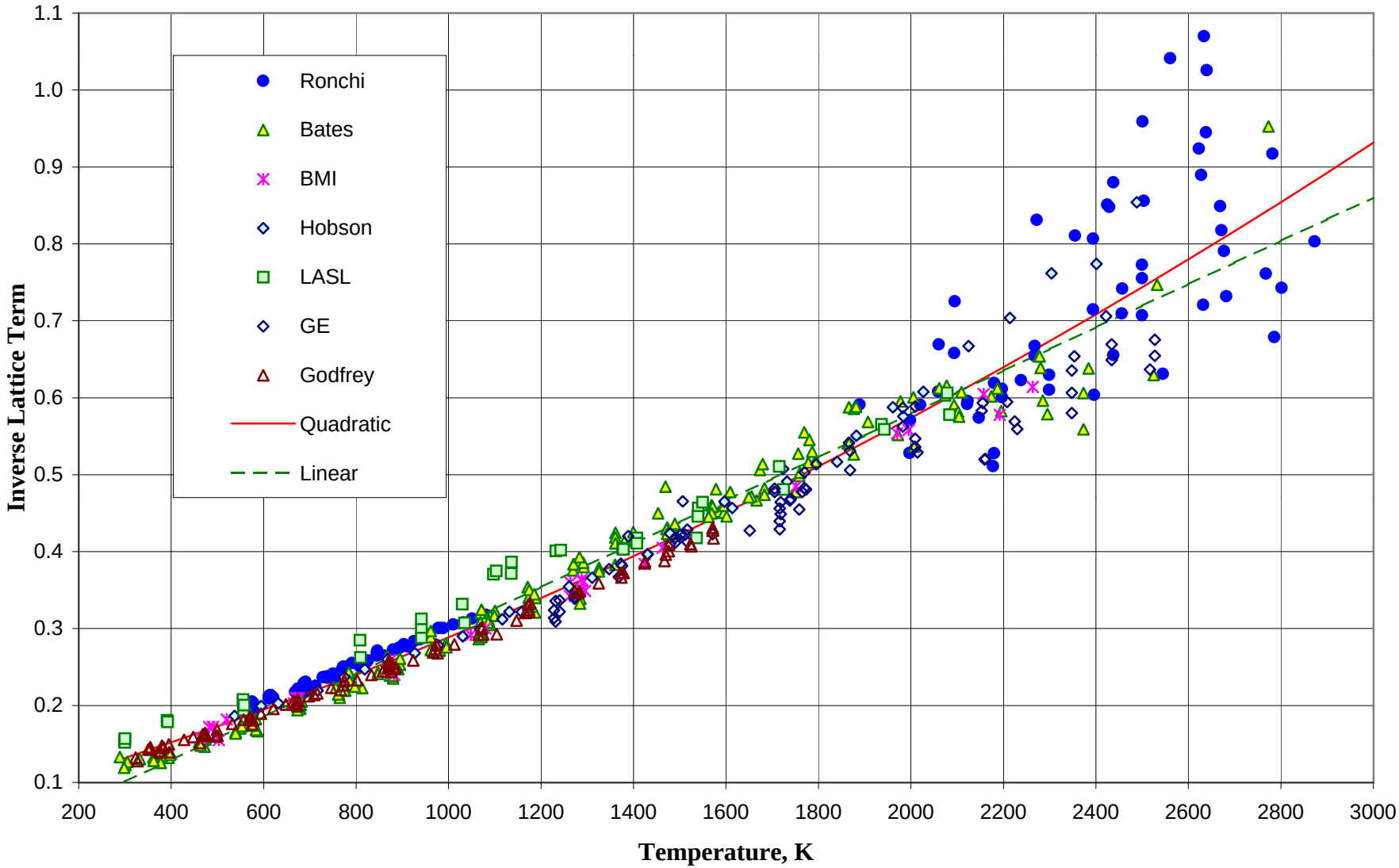

Figure 6 Comparison of Linear and Quadratic Lattice Terms in Thermal Conductivity Equations for 95% Dense UO_2

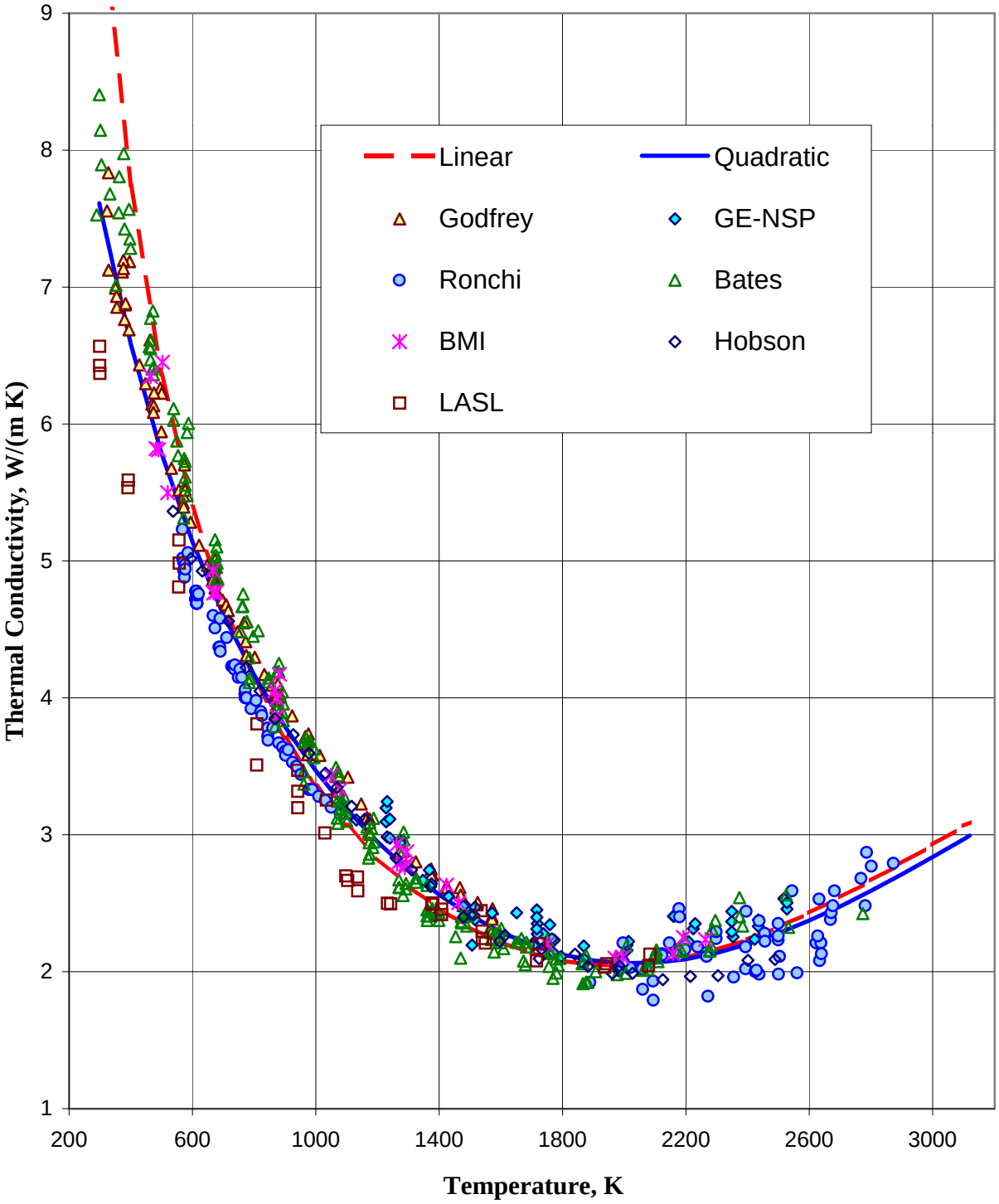

Figure 7 Percent Deviations from Equation with New Linear Lattice Term

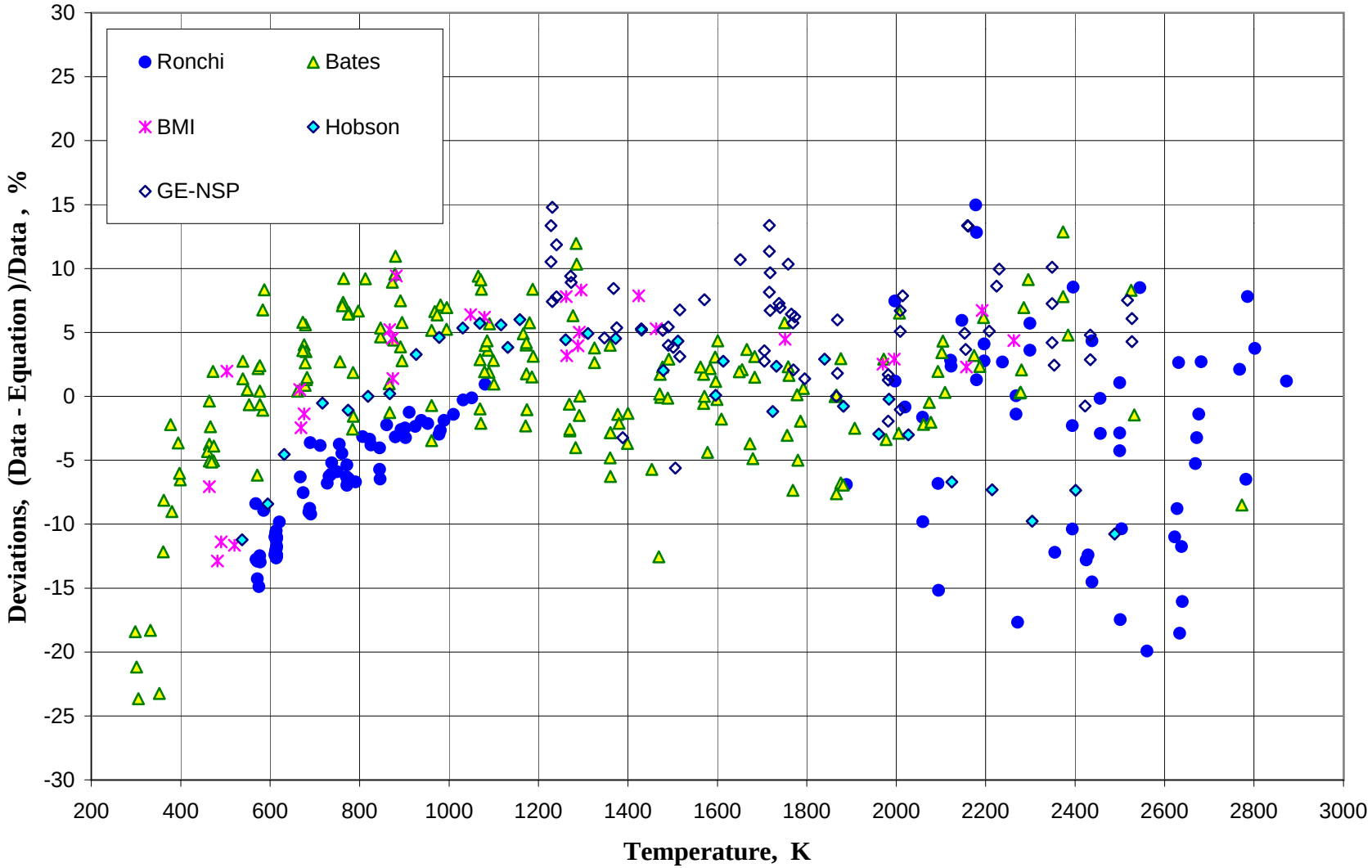

Figure 8 Percent Deviations from Equation with Quadratic Lattice Term

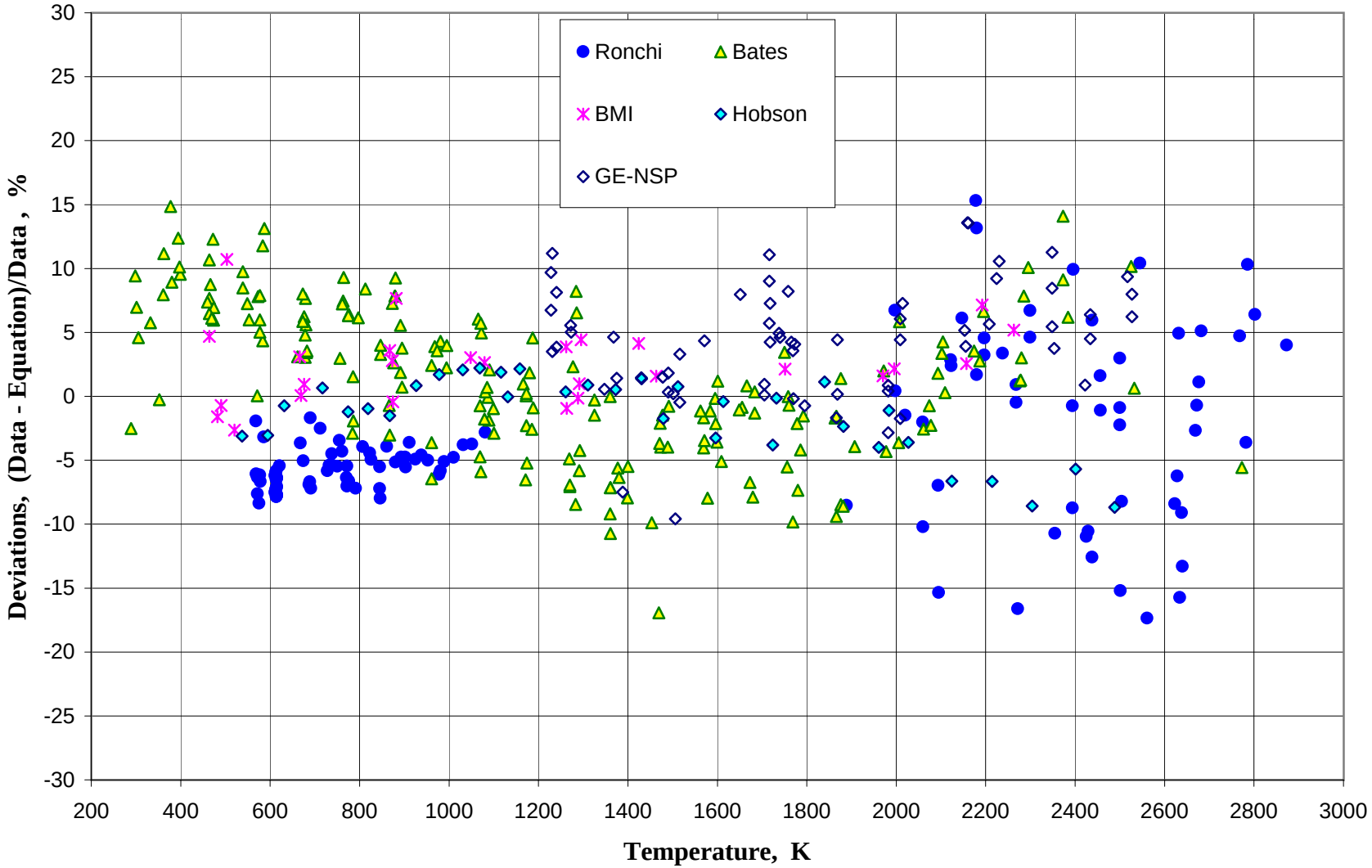

Figure 9 Thermal Conductivity of 95% Dense UO_2

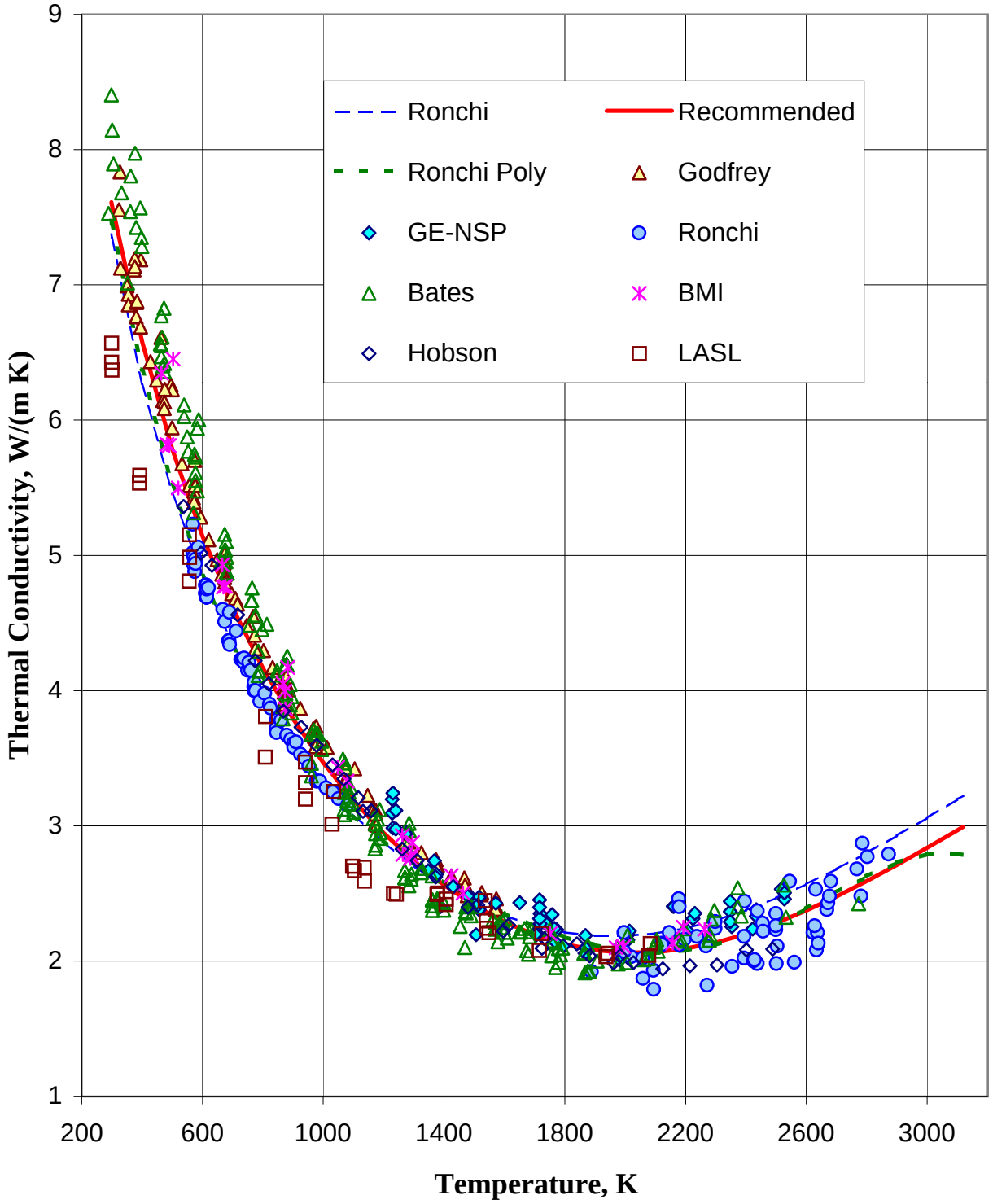

Figure 10 Percent Deviations from Equation of Ronchi

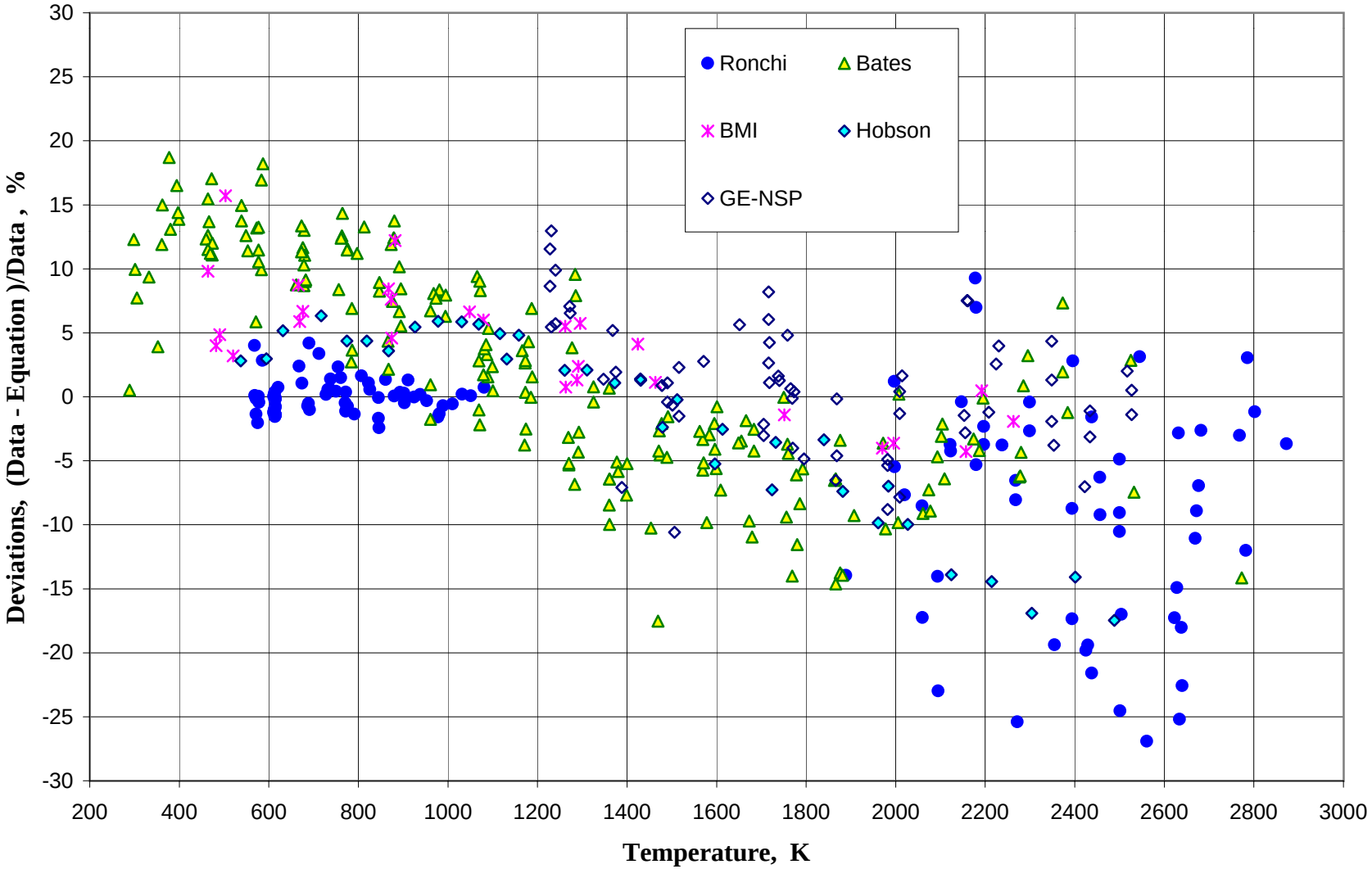

Figure 11 Comparison of Recommended Equation with Previous Recommendation and Data for 95% Dense UO_2

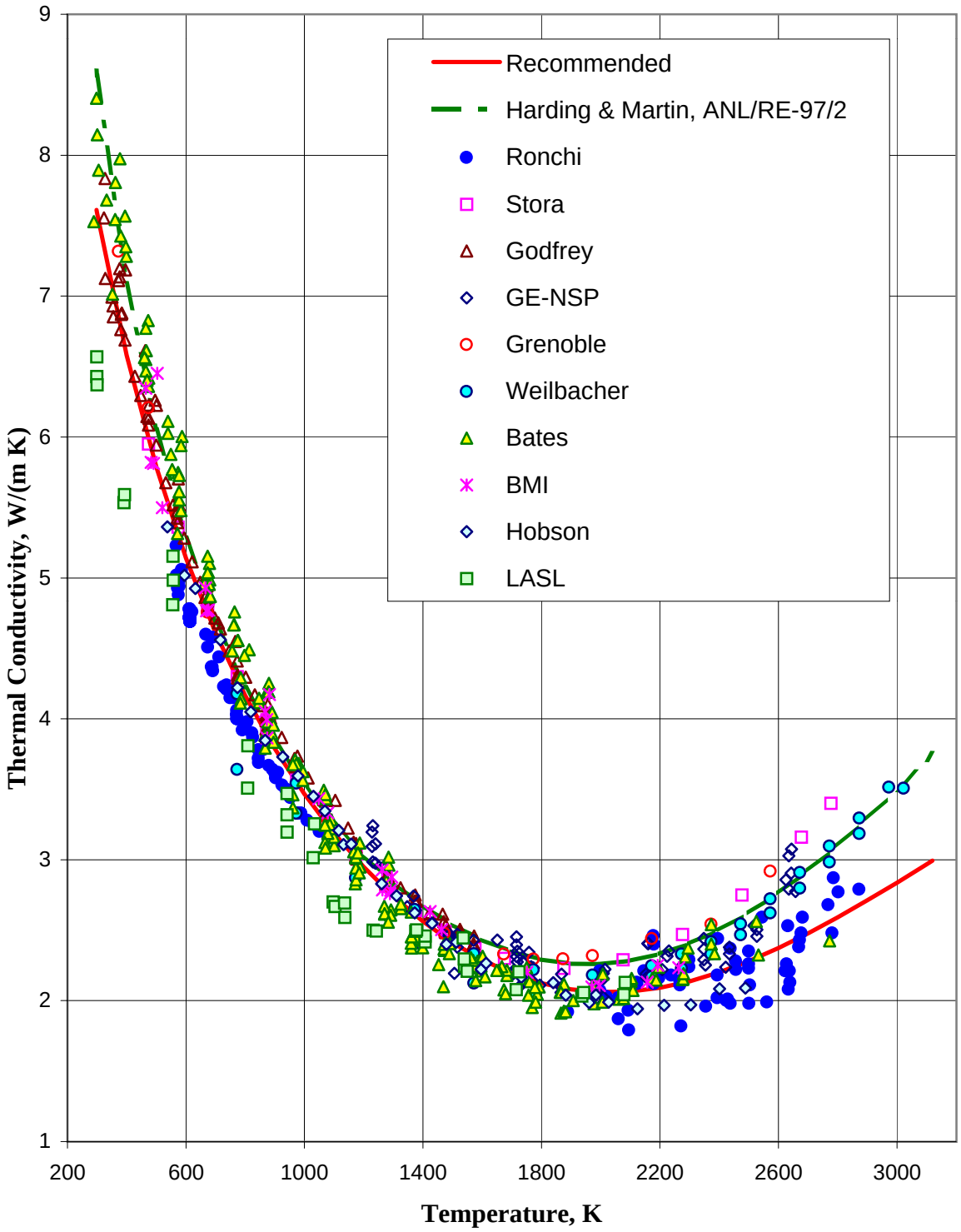

Figure 12 Percent Deviations from Recommended Equation

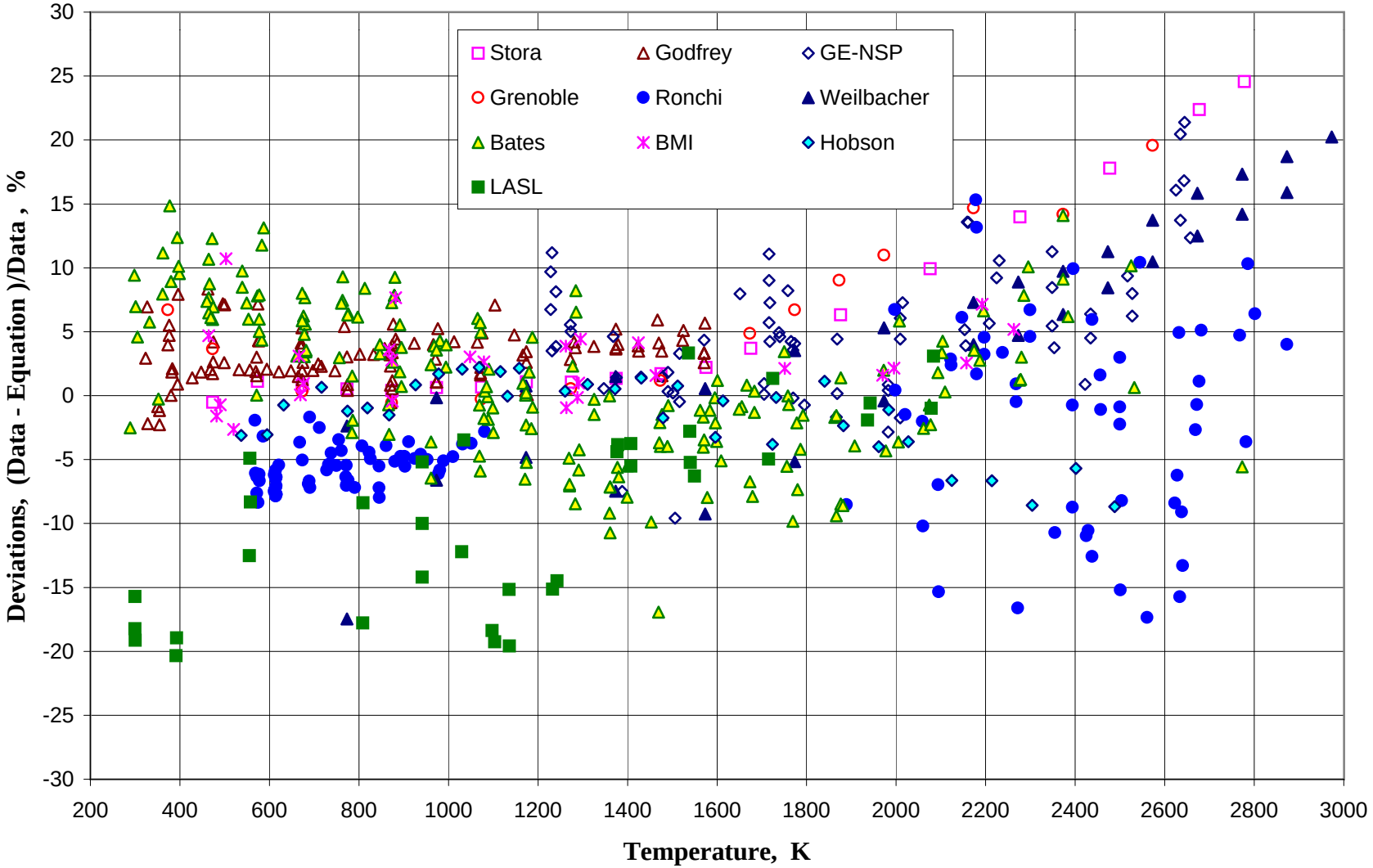

Figure 13 Comparison of Equations for the Thermal Conductivity of 100% Dense UO_2

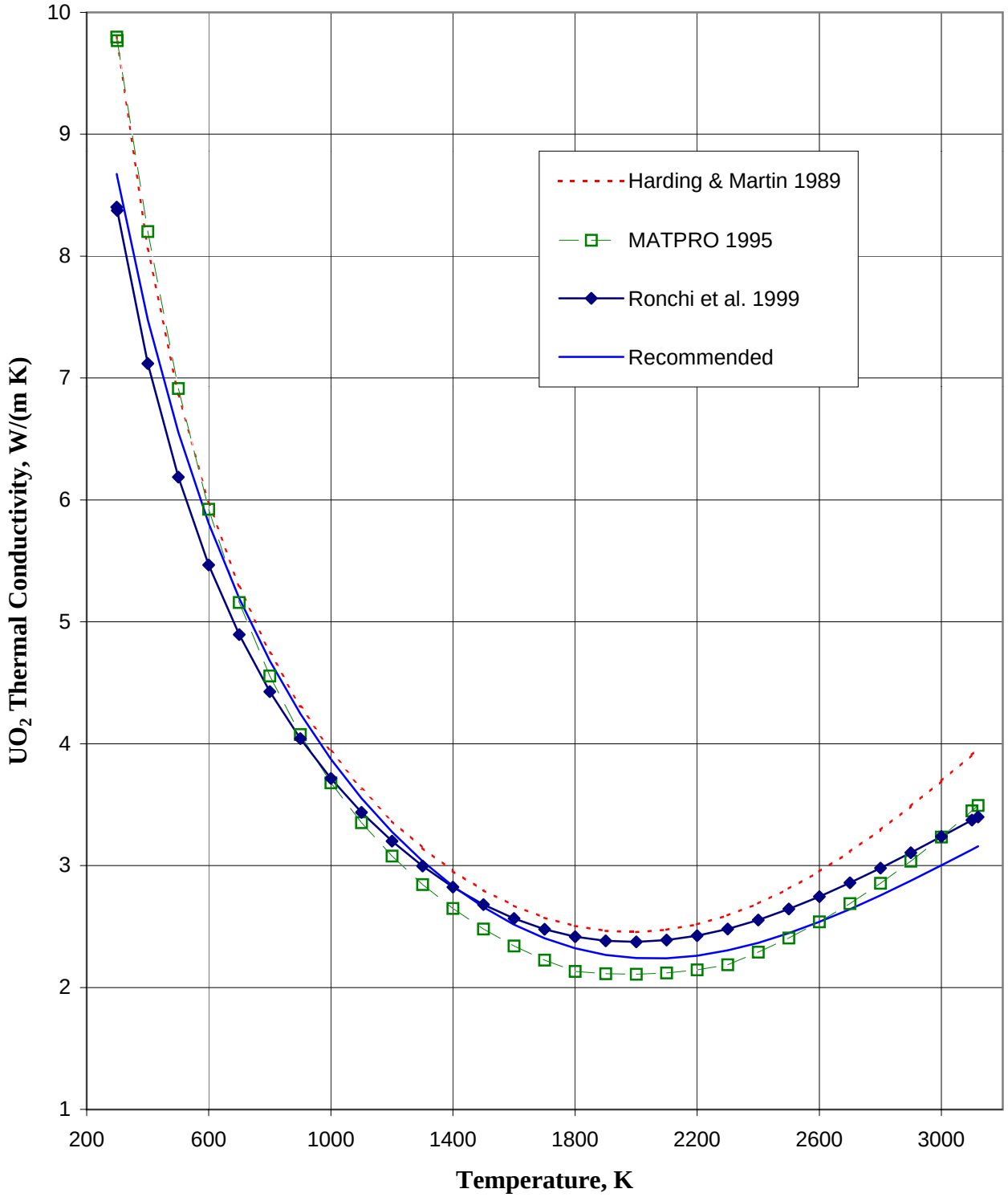

Figure 14 Percent Deviations from Recommended UO_2 Thermal Conductivities